Control of External Molecular Modes with Intense Light

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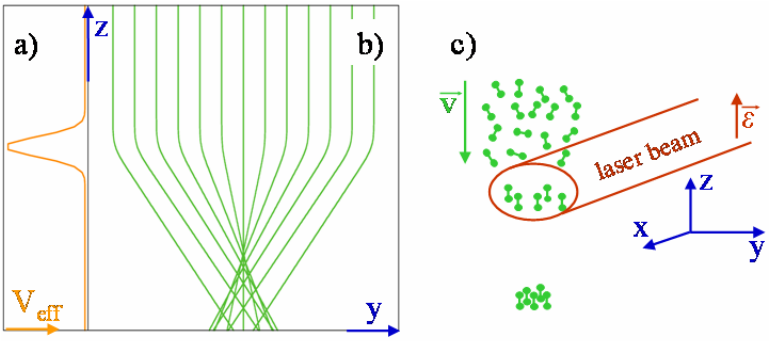
Abstract
Moderately intense laser fields can be used to excite rotationally broad, coherent superposition states with fascinating properties that may also be potentially useful. We illustrate the possibilities of forcing the three axes of an arbitrary polyatomic molecule to align along given three axes fixed in space, of preserving the field induced alignment after turn-off of the laser pulse, of forming superposition states that are perfectly aligned and fully isotropic periodically in time, and of focusing, collimating, steering, dispersing and reflecting molecular beams with light. Problems where we hope to identify new opportunities in the X-ray regime are noted.

Atom optics, the application of lasers to spatially manipulate atomic motions, has been one of the most active sub-disciplines of atomic physics, with applications ranging from atom holography to atom lithography. Unfortunately, the techniques of atomic manipulation with light cannot be extended to spatially manipulate molecular motions, due to the complex level structure and weak transition dipole elements of molecules. Success in the atomic domain nevertheless suggests that an analogous field of molecular optics could open a rich variety of new opportunities.

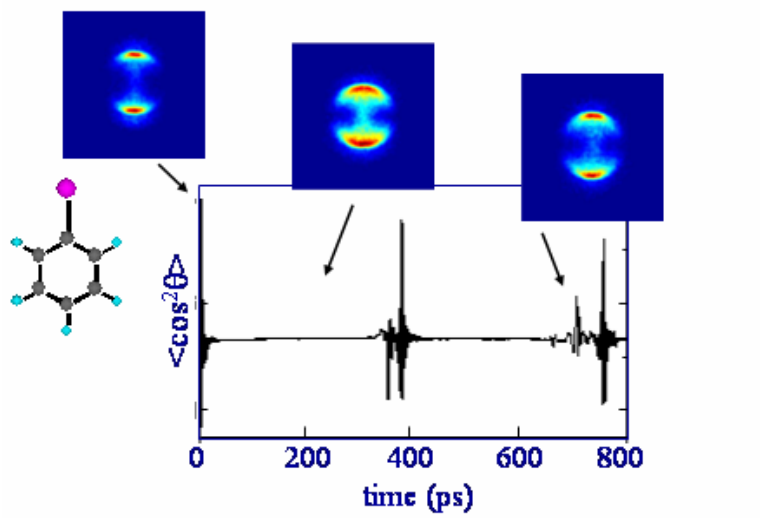
During the past few years it was shown, theoretically and experimentally, that general molecules can be focused and guided using a non-resonant, moderately intense laser field. Spatial manipulation in that case is based on the nonlinear interaction of the intense field with the quasi-static polarizability tensor, rather than on near-resonance interactions as in the atomic case. The spatial intensity profile of the strong field produces an effective well for the center-of-mass motion which accelerates the molecular trajectories toward the high intensity region and brings them to a focus in a predetermined point in space. The method is applicable to all molecules (and atoms), since all molecules are polarizable to some extend, and robust, since the deep well translates into weak sensitivity to velocity distribution and other aberrations. Molecular focusing generalizes to a field of molecular optics, with possible applications in nanoscale surface processing and molecular separation techniques.



Molecular focusing in moderately intense laser fields. (a) Quantum mechanically computed effective potential subject to which the center-of-mass motion evolves. (b) Classical trajectories describing the center of mass evolution subject to the laser induced potential. (c) Schematic illustration of the geometry along with definition of the axis system.
T. Seideman, Phys.Rev.A **56**, R17 (1997)

An important advantage of atom optics which the molecular optics scheme does not share, is the availability of both attractive and repulsive optical elements, obtained by red- or blue-detuning the laser frequency from resonance. The molecular optics scheme, by contrast, is based on attractive interactions alone. One of our purposes in the present talk is to examine the possibility of extending molecular optics to include repulsive optical elements and illustrate some of their potential applications

Related to intense-light molecular optics is the problem of molecular alignment in intense laser fields. From the formal view-point, both intense field alignment and photomanipulation of the center-of-mass motion rely on inhomogeneous field effects. Furthermore, both reduce in one limit to fully quantal cycles between two electronic states and in another to the nearly classical interaction of an electric field with a many level system. From a practical view point, the possibility of simultaneously aligning molecules and focusing their translational motion is intriguing, with potential applications in stereodynamics, gas-surface research, surface catalysis and material processing. It is clear, however, that to become useful, simultaneous focusing and alignment should be realized under field-free conditions. A second goal of this presentation is to suggest and examine a means of simultaneously focusing and aligning molecules in a field-free region of space.



Time dependent alignment induced by short pulses. Theory and experiments. The main panel shows the computed expectation value of $\cos^2\theta$ in the wavepacket during and after excitation of iodobenzene with a 2.6 ps pulse. The insets show three experimental images taken at different times during the evolution. E. Peronne, M. Poulsen, C. Bisgaard, H. Stapelfeldt & Tamar Seideman, Phys.Rev.Lett. **91**, 043003 (2003).

Conventional alignment is a one-dimensional concept; a uniaxial field can only define one direction in space. It will be shown that by means of an elliptically polarized field it is possible to force the three axes of an arbitrary polyatomic molecule to align along given three axes fixed in space. Potential applications of three-dimensional alignment, ranging from control of charge transfer reactions in solutions through guided molecular assembly for molecular devices to quantum storage, will be discussed.